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Waste Paper for Recycling: Overview and Identification of Potentially Critical Substances

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Abstract

Paper product manufacturing involves a variety of chemicals used either directly in paper and pulp production or in the conversion processes (i.e. printing, gluing) that follow. Due to economic and environmental initiatives, paper recycling rates continue to rise. In Europe, recycling has increased by nearly 20% within the last decade or so, reaching a level of almost 72% in 2012. With increasing recycling rates, lower quality paper fractions may be included. This may potentially lead to accumulation or un-intended spreading of chemical substances contained in paper, e.g. by introducing chemicals contained in waste paper into the recycling loop. This study provides an overview of chemicals potentially present in paper and applies a sequential hazard screening procedure based on the intrinsic hazard, physical-chemical and biodegradability characteristics of the substances. Based on the results, 51 substances were identified as potentially critical (selected mineral oils, phthalates, phenols, parabens, as well as other groups of chemicals) in relation to paper recycling. It is recommended that these substances receive more attention in waste paper.

Keywords: Hazardous substances; Paper; Priority pollutants; Recycling; Waste management

Abbreviations

BBP: Benzyl butyl phthalate

BPA: Bisphenol A

CAS: Chemical Abstracts Service

CEPI: Confederation of European Paper Industries

DBP: Dibutyl phthalate

DEHP: Diethylhexyl phthalate

DIBP: Diisobutyl phthalate

DIPN: Diisopropyl naphthalene

EDCs: Endocrine Disrupting Chemicals

EFSA: European Food Safety Authority

EuPIA: European Printing Ink Association

FDHA: Swiss Federal Department of Home Affairs

NIAS: Non-Intentionally Added Substances

PCBs: Polychlorinated biphenyls

PBT: Persistent, Bioaccumulative and Toxic

vPvB: very Persistent and very Bioaccumulative

ZELLCHEMING: Vereins der Zellstoff- und Papier-Chemiker und –ingenieure (German for: Association of Chemical Pulp and Paper Chemists and Engineers).

1. Introduction

Paper recycling is one of the most well-established recycling schemes applied to waste materials today. Recycled paper is an integral part of paper and pulp production, with estimated utilisation for recycling in Europe of about 72% in 2012 (an increase of 20% from 2000) (CEPI, 2013a). In addition to recycled paper being an important raw material for the paper industry (CEPI, 2013b), it has also been demonstrated in several studies that paper recycling may offer significant environmental benefits in a lifecycle perspective (Laurijssen et al., 2010; Villanueva and Wenzel, 2007). Thus, paper recycling may be regarded as beneficial from both a resource and an environmental perspective and should be promoted as much as possible. However, increasing concerns related to the presence of potential harmful chemical substances in paper have been voiced within recent years (e.g. Biedermann et al., 2011b; Liao and Kannan, 2011; Pivnenko et al., 2013), for example in relation to the migration of chemicals from packaging materials into food (e.g. Begley et al., 2008; Biedermann et al., 2013; Gärtner et al., 2009; Lorenzini et al., 2013). While further increasing paper recycling rates can undoubtedly be achieved in Europe, the quality of the waste paper may ultimately decrease as more and more "marginal" paper fractions are collected for recycling and the contents of harmful substances in paper thereby increase. A systematic overview of the chemical substances potentially present in waste paper for recycling is therefore needed to provide a basis for further evaluation of the quality of waste paper as a resource, and ultimately also to maintain consumer acceptance of recycled paper in general.

Paper production and manufacturing operations generally consist of the following two phases: i) paper and pulp production by the paper industry (i.e. different quality grades of paper) and ii) paper product manufacturing by separate industries (e.g. periodicals, packaging materials, books, etc.). Chemicals in waste paper may originate from a wide range of sources, namely intentionally added (i.e. additives, inks, pigments, glues, etc.), part of a reaction and/or biodegradation or added during the use phase of the paper or during the waste management phase (e.g. cross-contamination from other waste materials during collection). Chemicals are added in order to improve the production process itself and the quality or functionality of the final product. Starting with paper production, chemicals are introduced through the use of synthetic additives, which include retention aids, sizing agents, coatings, biocides, synthetic binders, etc. Synthetic additives represent slightly more than 1% v/v of raw materials used in paper production (ZELLCHEMING, 2008), the largest share of which (90% v/v) are functional additives (Moench and Auhorn, 2002) intended to be retained in the paper product. The next step, where the paper is converted into a final product, may include printing, dyeing, addition of adhesives and labels, etc. During the processing, chemicals may dissolve and be removed via wastewater, volatilize and be released to air or remain in the solid matrix and thereby be present in newly manufactured paper products. When waste paper is added to the process, this may potentially introduce new substances from the use and waste management phase. Knowing which potential partitioning a given chemical (or group of chemicals) will follow is vital for identifying potentially critical substances which may end up being concentrated in the fibres and be reintroduced into consumer products.

Recent studies have demonstrated that paper and paper products may contain high numbers of chemical substances (BMELV, 2012; Bradley et al., 2008), most of which can be associated with the printing industry, where more than 7,000 chemicals may be used in food-packaging ink production alone (EuPIA, 2012).Nevertheless, very little quantitative information is available regarding the presence of specific substances in paper products or waste paper potentially sent to recycling. Most existing studies target a specific group of chemicals or paper products (e.g. Becerra and Odermatt, 2012; Geens et al., 2012; Song et al., 2000; Trier et al., 2011), and attempting to identify every single chemical present in paper has proved to be challenging (BMELV, 2012).

Although specific regulations covering paper food packaging do not exist, European legislation on items (i.e. plastics, metal, paper, etc.) brought into contact with food prevents the use of chemicals that could migrate into foodstuffs and adversely affect human health, as well as the quality and nature of food (EC, 2004). This legislation covers paper packaging produced from virgin fibres, but when paper is recycled, the producers may not be aware of the presence of any specific chemicals added throughout the lifecycle of the paper. Consequently, the paper industry, and the final output paper quality, is affected by the presence of chemicals in the recycled paper, e.g. chemicals introduced during the use phase or via paper products from other countries. In 2012, more than 5 million tonnes of paper (approx.11% of recycled paper) was imported into Europe from the USA, Russia, Brazil, Canada, etc. for paper product manufacturing (CEPI, 2013a).

Without a comprehensive overview of which chemical substances should be prioritised in relation to paper, and which substances should ultimately be avoided, it may not be possible in the future to ensure both high recycling rates and at the same time a high quality of the paper products based on recycled fibres. As direct and substance-by-substance analysis is not practically feasible, a systematic screening of un-problematic chemicals is needed, in order to identify those substances which may be considered most problematic and critical for the future recycling of paper.

The overall goal of this study is to provide a basis for systematically addressing the recyclability of waste paper with respect to the potential presence of hazardous substances. The specific objectives are: i) based on existing literature, to compile a list of chemical substances potentially applied in paper production and paper product manufacturing, as well as chemicals identified directly in paper, ii) based on a sequential hazard screening procedure to identify the most critical chemicals from this list based on their harmfulness, physical-chemical properties and biodegradability and iii) to evaluate potential implications related to the management of paper waste and paper recycling.

2. Methodology

2.1 Data sources for chemicals in paper

Information about chemical substances, used in either paper production or paper conversion, as well as chemicals identified in actual paper product flows, was obtained from a range of data sources. Chemicals used in pulp and paper production were obtained from national product registries (KEMI, 2014; SPIN, 2013) and scientific assessments (Riskcycle, 2013; ZELLCHEMING, 2008), as well as inventory data provided by the European Food Safety Authority (EFSA) (EFSA, 2012a). Substances used by the printing industry were obtained from a recent Danish report (Miljøstyrelsen, 2011a), an inventory list of the European Printing Ink Association (EuPIA) (EuPIA, 2012) and recent regulation issued by the Swiss Federal Department of Home Affairs (FDHA) (FDHA, 2005). Although data obtained for paper printing could not be isolated from the printing of other materials, the European printing industry belongs to a forest-based industrial sector, and the share of paper in the printing industry is substantial. All of the abovementioned data sources predominantly reflected European industry and research; this was not due to any selection of sources, but rather reflected availability of state-of-the-art information and level of detail provided. No information could be found related specifically to chemicals used in adhesives, so these were therefore only indirectly included in the study as part of the analytical literature reviewed. Additionally, relevant scientific literature addressing the composition of paper, paper products or waste paper was reviewed. While the aim was not to provide an exhaustive review of all available literature, the focus was placed on recent literature in order to relate any findings as best as possible to the current technological scope of the paper industry. No geographical scope was applied to the selected studies, as paper is a commodity traded on the global market with high volumes of paper, paper packaging and waste paper being imported and exported on a yearly basis. In total, 25 scientific studies were reviewed.

Where available the concentrations of substances mentioned in the literature are also provided. See Table 1 for a complete list of the data sources used in this paper.

Based on the abovementioned combination of information sources, a compilation of almost 10,000 chemical substances was obtained once duplicates were removed. To avoid ambiguity and potential double-counting, only chemicals (or groups of chemicals) which could be assigned a valid CAS (Chemicals Abstracts Service) registry number were included in the study.

2.2 Criteria for identifying potential priority chemicals

With the aim of identifying potentially critical chemicals that should be prioritised in relation to paper recycling, a screening selection procedure was applied for those that may be considered most harmful, most likely to be associated with paper fibres (and not volatilise or be released into the water phase during re-pulping) and the most persistent in the environment. The procedure involved the following four steps: 1) compiling an inventory of chemicals that may be used in the paper and printing industries or which may have been identified in paper (corresponding to the list of about 10,000 substances mentioned above), 2) identifying potentially harmful chemicals, 3) identifying chemicals primarily associated with solids (i.e. paper fibres) and 4) identifying chemicals characterised as not readily biodegradable. Steps 1) through 4) were carried out consecutively, thereby filtering out less problematic substances in relation to paper recycling. The remaining list of chemicals therefore represented substances that should be prioritised in future scenarios characterising paper and addressing paper recycling. See Figure 1 for an illustration of the procedure.

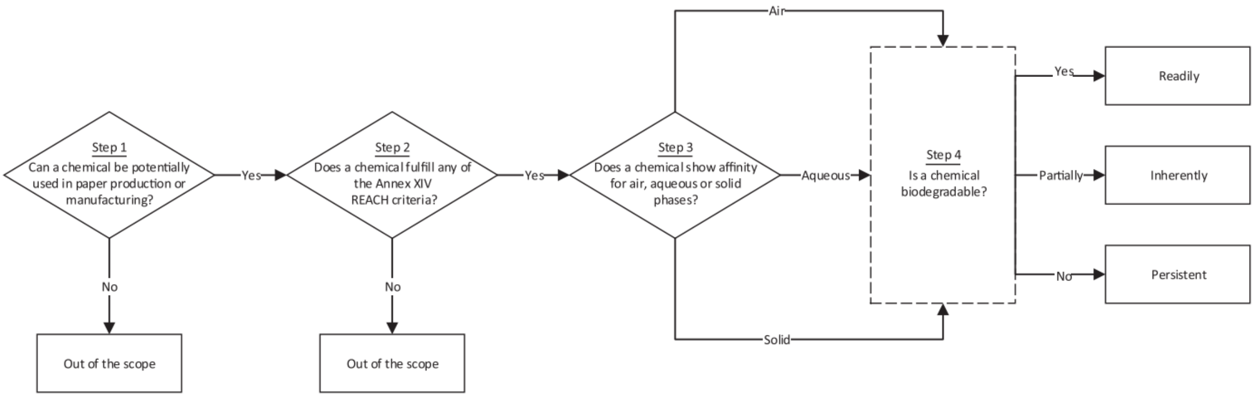

Figure 1. Schematic representation of the methodology applied in selecting relevant chemicals of interest.

In Step 2), chemicals were selected in accordance with Annex XIV of European REACH Regulation (EC, 2006) and according to the following criteria: i) substances classified in hazard class "carcinogenicity" (categories 1A and 1B, Carc. 1A or 1B) (EC, 2008), ii) substances classified in hazard class "germ cell mutagenicity" (categories 1A and 1B, Muta. 1A or 1B) (EC, 2008), iii) substances classified in hazard class "reproductive toxicity" (categories 1A and 1B, Repr. 1A or 1B) (EC, 2008), iv) substances classified as "Persistent, Bioaccumulative and Toxic (PBT)" (according to Annex III in (EC, 2006)), v) substances classified as "very Persistent and very Bioaccumulative (vPvB)" (according to Annex III in (EC, 2006)) and vi) substances characterised as "Endocrine Disrupting Chemicals (EDCs)" (WHO, 2002) for which scientific evidence of possible serious effects on human health and/or the environment could be found. The chemicals selected based on Step 2 included all substances fulfilling at least one of the abovementioned criteria. Only substances with sufficient information available were selected in Step 2; in other words, those with non-published or incomplete hazard assessments were not included. In practice this means that the number of chemicals finally selected in this study might be underestimated, as future hazard assessments of chemicals included in inventory list (Step 1) may reveal additional substances fulfilling the Step 2 criteria.

Step 3) was based on the methodology described by Baun et al. (Baun et al., 2006), where partitioning between phases is based on the potential of a given chemical to be adsorbed to solids, to volatilise or to remain dissolved in the aqueous phase. Influence of particular paper production processes (pulping, coating, drying, etc.) on phase distribution of substances is out of the scope of the present work and was not considered.

In Step 4) of the screening process, chemicals were assessed in accordance with their biodegradability and then classified into persistent, inherently and readily biodegradable. Classification was based either on the Biowin models 3 and 5, included in EpiSuite 4.1 (U.S. EPA, 2013) with cut-off values as presented by Baun et al. (Baun et al., 2006), or on scientific literature providing experimental biodegradation results. Relevance of biodegradability of chemicals to particular processes in paper recycling was not established, as the variations associated with different steps of the paper lifecycle (i.e. paper production, manufacturing, use, waste paper collection, re-processing, etc.) are potentially large.

3. Results and Discussion

3.1 Overview of substances

Figure 2 presents material flows of European paper recycling, indicating points where chemicals are introduced into the loop. Most non-fibrous materials are introduced in the paper production step, but they are almost entirely represented by non-hazardous naturally occurring substances such as clay, $CaCO_3$ and starch. No quantitative data were available regarding chemicals added non-intentionally into the loop (i.e. Non-Intentionally Added Substances, NIAS). An overview of each of the sources contributing to the final list in Step 1 is presented in Table 1. Although the paper industry uses high volumes of chemical substances (Figure 2), it is evident that a much higher variety of chemicals associated with paper products derives from printing (Table 1).

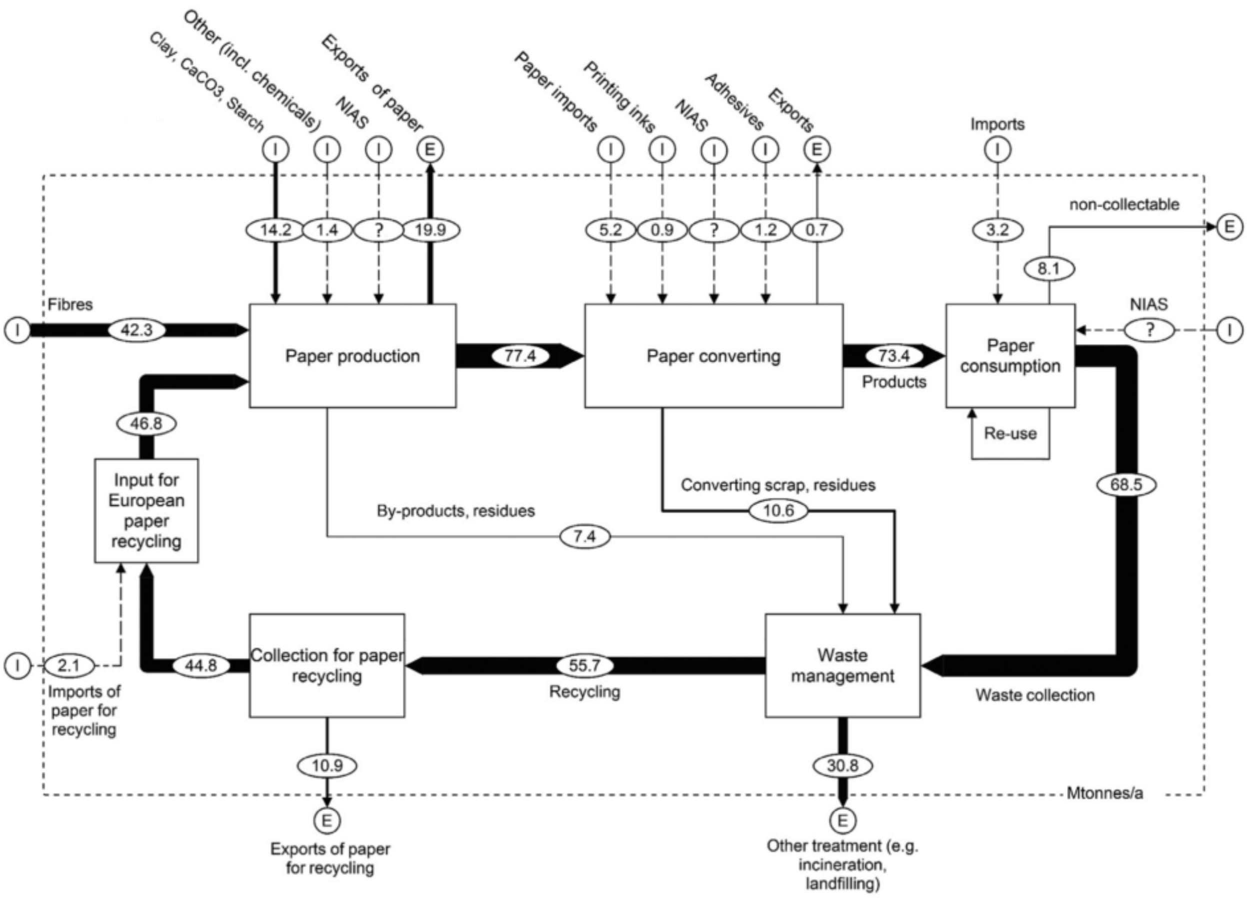

Figure 2. Material flow of the European paper recycling loop. Dotted lines indicate points where chemicals are introduced (Based on (CEPI, 2013a, 2013b; EUPIA, 2013; FEICA, 2008.; ITC, 2014) and personal communication with the Confederation of European Paper Industries (CEPI)).

Due to the large number of chemicals identified in Step 1, attributing to each of them a potential use by industry is practically impossible. Nevertheless, most of the substances used in paper production can be attributed to fillers, binders, retention aids, wet/dry-strength agents, coaters, biocides, dispersers, etc. (ZELLCHEMING, 2008). In the printing industry the vast majority of chemicals are used as solvents, dyes, inks, pigments, binders, curing agents and photoinitiators, plasticisers, surfactants, etc. (Miljøstyrelsen, 2011a) Only a small fraction (157) of the almost 10,000 substances could be identified in Step 2. Figure 3a presents the distribution of substances on Step 2 list in accordance with their use by industries throughout the lifecycle of paper. Only 10 of the chemicals are used exclusively in paper production (mainly biocides). Conversely, 133 chemicals were attributed to the printing industry, most of which are solvents and polymeric resins employed in inks, pigments and dyes. Chemicals which could not be attributed either to paper production or to the printing sector (14) could potentially be by-products or contaminants introduced into the production cycle through recycled paper.

Table 1. Data sources used in the study and their quantitative contribution to Step 1 list of chemicals.

Source	**Number of chemicals**	**Description**	**Industry**	**Reference**
Literature	348	Scientific literature providing analytical data on the identification or quantification of chemicals in paper and/or board	Paper and paper product manufacturing, and NIAS	(Biedermann et al., 2013, 2010; Binderup et al., 2002; BMELV, 2012; Bradley et al., 2008; Castle et al., 1997a, 1997b; Fierens et al., 2012; Gehring et al., 2004; LeBel et al., 1991; Liao and Kannan, 2011; Miljøstyrelsen, 2011b, 2003a, 2003b; Ozaki et al., 2004; Parry, 2001; Petersen et al., 2013; Poças et al., 2010; Riber et al., 2009; Rotter et al., 2004; Sipiläinen-Malm et al., 1997; Storr-Hansen and Rastogi, 1988; Sturaro et al., 2006; Vinggaard et al., 2000; Zheng et al., 2001)

Danish product register	75	Chemicals used in preparations of articles. Danish industry for pulp, paper and paper products	Paper and paper product manufacturing	(SPIN, 2013)
Swedish product register	144	Chemicals used in preparations of articles. Swedish industry for pulp, paper and paper products	Paper and paper product manufacturing	(KEMI, 2014)
Danish Environmental Protection Agency	415	Inventory of chemicals used by the Danish printing industry	Paper product manufacturing*	(Miljøstyrelsen, 2011a)
RiskCycle	12	Database of chemical additives used in paper production	Paper manufacturing	(Riskcycle, 2013)
ZELLCHEMING	44	Chemical additives for the production of pulp and paper	Paper manufacturing	(ZELLCHEMING, 2008)
EFSA	223	Chemicals currently used in the manufacture of paper and board	Paper manufacturing	(EFSA, 2012a)
FDHA	4575	Chemicals permitted to be used in the manufacture of	Paper product manufacturing*	(FDHA, 2005)

		packaging inks		
EuPIA	3858	Inventory list of chemicals used in the manufacture of food packaging inks	Paper product manufacturing*	(EuPIA, 2012)
TOTAL:	9694	-	-	-

*not limited to paper matrix

Chemicals on the Step 2 list were assessed in relation to their potential partitioning between the air, aqueous and solid phases. It is evident from Figure 3b that most of the chemicals either have a higher affinity for a solid matrix (51) or remain dissolved in the water phase (51). Sixteen of the chemicals on the list are relatively volatile and could potentially escape through volatilisation in the production process, while an additional 22 substances on the list are inorganic, and although two of them (i.e. mercury and carbon disulphide) may partially volatilise, the distribution of the rest will depend very much on specific conditions in the paper processing stages (e.g. pH, redox conditions, presence of organic matter, etc.) and are difficult to predict. Chemicals remaining in the solid matrix are of particular interest in terms of paper recycling.

In the following step (Step 4) the biodegradability of the previously identified chemicals was assessed. As presented in Figure 3c, most of the substances that showed affinity for the solid phase were characterised as persistent (24), while the 27 remaining chemicals could be classified as inherently (12) and readily biodegradable (15).

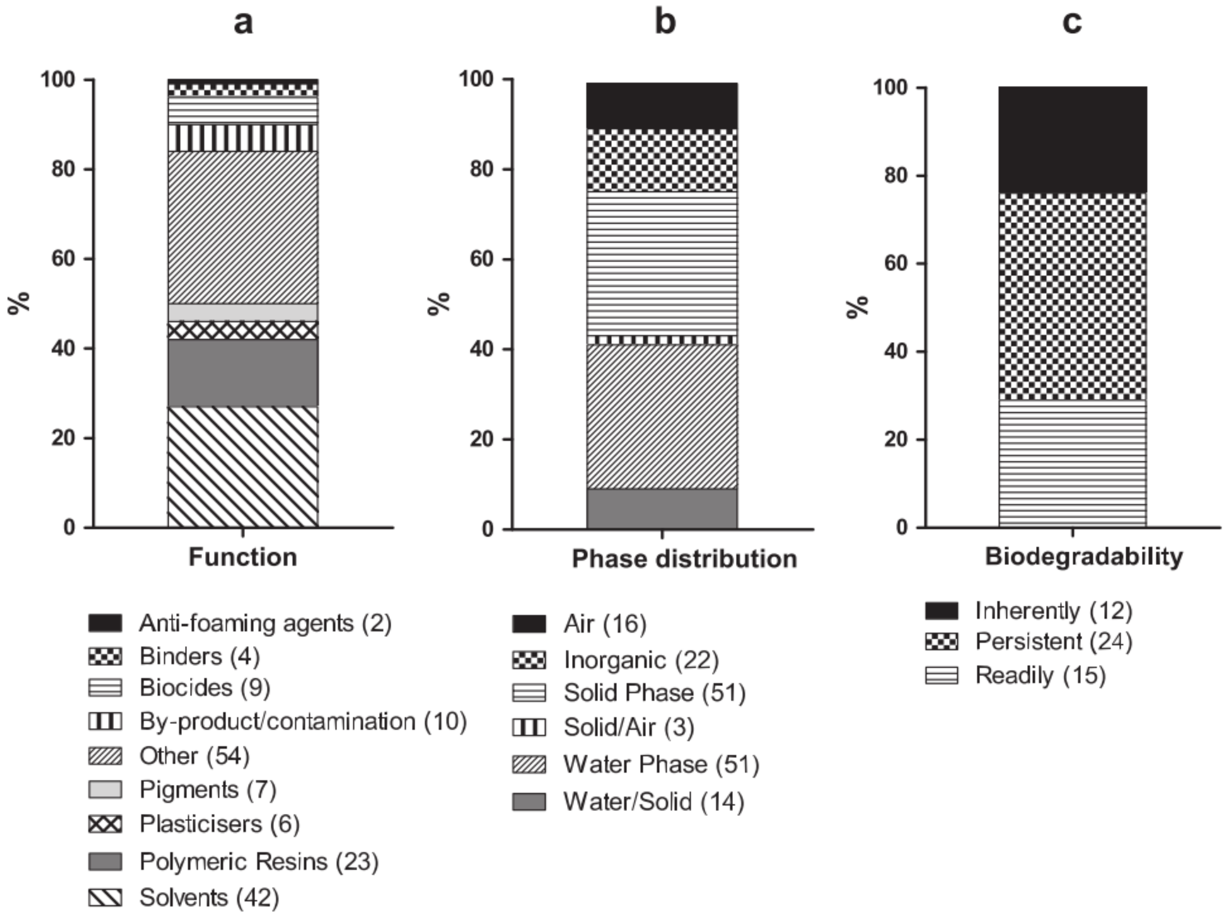

Figure 3. Distribution of the use of chemicals from Step 2 list (3a); phase distribution of chemicals, i.e. Step 3 list (3b); biodegradability of chemicals associated with the solid phase, i.e. Step 4 list (3c).

While the list of potential priority chemical substances may not be exhaustive (as the screening can only be based on available information about substances), it nevertheless clearly indicates that concerns regarding their presence in paper and their potential accumulation in the paper lifecycle may be pertinent to a relatively small number. The list therefore forms a systematic basis for further research in relation to paper characteristics and paper recycling. The 157 substances identified in Step 2 are grouped in the following according to their chemical structure and then discussed in more detail. Individual tables listing each chemical substance according to these groups can be found in the Supplementary Materials (Tables S1-S6).

3.2 Mineral oils

The commonly used term "mineral oils" refers to a mixture of components which originate from crude oil refining processes. Mineral oils mainly contain straight and branched open-chain alkanes (paraffins), alkylated cycloalkanes (naphthenes) and aromatic hydrocarbons (EFSA, 2012b), and their final composition will depend largely on the initial composition of the crude oil, as well as the refinery treatment (e.g. alkylation, hydro-treatment, cracking, extraction, etc.). Although the Joint FAO/WHO Expert Committee on Food Additives (JECFA) recently withdrew previously established acceptable daily intakes in relation to mineral oils (JECFA, 2012), the JECFA assessment refers to highly-refined mineral oils free from aromatic hydrocarbons. On the other hand, paper products were shown to contain technical-grade mineral oils which may include aromatic hydrocarbons (Biedermann and Grob, 2010). Grob et al. (Droz and Grob, 1997) found that, at least initially, printing inks (solvents in particular) used in paper products are the main source of mineral oils in paper. Further studies have also posited that mineral oils may derive from recycled waste paper (Biedermann and Grob, 2010; Biedermann et al., 2011b).

Out of the 157 chemicals included in the Step 2 list, 49 were classified as mineral oils (Table S1 (Supplementary Material)) and characterised as carcinogens, while some are also mutagenic substances. The chemicals presented herein are not single substances but rather mixtures of substances containing various hydrocarbons. Being mixtures rather than single substances, mineral oils can be used in a variety of applications in the industry, i.e. from solvents and as the basis for polymeric resins through to lubricants and cleaning agents for machinery (EFSA, 2012b; Miljøstyrelsen, 2011a). Most of the scientific studies currently available focus on mineral oil content in paper used for food packaging (e.g. (Biedermann and Grob, 2010; Biedermann et al., 2011a, 2011b; Droz and Grob, 1997)), as migration into foodstuffs remains one of the most important sources of consumer exposure (EFSA, 2012b). As they are hydrophobic substances, mineral oils may not be removed in water-based processes of paper recycling (i.e. pulping, deinking, washing), remain in the solid matrix and have a high chance of persisting in the recycling process and being reintroduced into newly manufactured products (BMELV, 2012). Such a scenario is unlikely for some of the lighter mineral oils, which are expected to escape due to volatilisation in e.g. paper drying step. A recent study (BMELV, 2012) showed that the deinking process reduces insignificantly the concentration of mineral oils, while paper drying is the main process for their removal (around 30% (Biedermann et al., 2011b)) – still resulting on average in 340 mg/kg ($<C_{24}$) in unprinted food-packaging board produced (Biedermann and Grob, 2012). One study showed that even the presence of a barrier (e.g. plastic foil) may not always prevent the migration of mineral oils from packaging into a food product (Fiselier and Grob, 2012), and a biodegradation assessment has shown that a significant number of mineral oils (15) can be classified as persistent, making bioaccumulation relevant for some.

Due to the diversity of mineral oils, and the fact that they are mixtures, identifying and quantifying single constituents (as the ones presented in Table S1 (Supplementary Material)) is practically impossible. As a result, mineral oils are analysed instead as sum parameters (e.g. the Hydrocarbon Oil Index), with fractioning based on the number of carbon atoms in the chemical (Droz and Grob, 1997; Pivnenko et al., 2013) or fractioning between mineral oil saturated and aromatic hydrocarbons (Biedermann and Grob, 2010; Biedermann et al., 2011b). The study conducted by Pivnenko et al. (2013) showed the presence of mineral oils in all the analysed waste paper fractions, with the highest concentrations (up to 1,800 mg/kg) identified in newspapers and tissue paper. Similarly, among waste paper materials fed into the German recycling loop, newspapers were identified as the main source of mineral oils (BMELV, 2012). Their presence in newspapers can be attributed to solvents and processes used in cold off-set printing (Biedermann and Grob, 2010), while mineral oils in tissue paper may indicate the introduction of chemicals during the product's life span and waste management. Both studies mentioned above (BMELV, 2012; Pivnenko et al., 2013) present relatively stable concentrations of mineral oils in a variety of board products, potentially indicating a contribution made by newspaper recycling.

3.3 Phthalates

Most phthalates are used as plasticisers in the preparation of printing inks, lacquers and dispersion glues (BfR, 2007; CDC, 2009), though they can also be used as softeners in tissue paper (Miljøstyrelsen, 2003a). From the Step 2 list, seven phthalates were identified (Table S2 (Supplementary Material)), the majority of which are classified as EDCs, while reproductive toxicity is also attributed to some. Phase distribution assessment (Step 3) revealed that phthalates may be retained in the paper and pulp solid matrices and could potentially follow the production process until the final product. Benzyl butyl phthalate (BBP), Dibutyl phthalate (DBP) and Diethylhexyl phthalate (DEHP) are classified as persistent, according to the criteria used. Table S2 (Supplementary Material) reveals the range of concentrations of phthalates quantified in paper, with Diisobutyl phthalate (DIBP) reaching the highest concentrations (up to 120 mg/kg). A study conducted by a German authority (BMELV, 2012) showed that phthalates were mainly present (up to 35 mg/kg) in board, waste paper from offices, specialty paper and papers containing relatively high amounts of glue. In contrast, newspapers, magazines and advertisements contained almost one order of magnitude lower phthalate concentrations (BMELV, 2012). These results could potentially indicate adhesives as the main source of phthalates in paper for recycling.

Experimental results involving four separate recycling facilities producing board for food packaging indicated that in the recycling process, phthalates have a high affinity for paper fibres, moving through the production line and then into final products (BMELV, 2012). Particularly, the study showed that DIBP, DBP and DEHP have a tendency (on average) to accumulate in board produced from recycled paper. On the other hand, the same study showed that virgin fibre-based board contained phthalates in lower concentrations (<0.2 mg/kg) well below one order of magnitude.

3.4 Phenols

Among the 157 chemicals in Step 2, eight were identified as phenols (Table S3 (Supplementary Material)), all of which fulfilled the EDCs criteria. The use of phenols in the paper industry varies significantly; for example, Bisphenol A (BPA) is used as a developer in thermal paper and pentachlorophenol as a biocide in paper production (Mendum et al., 2011; ZELLCHEMING, 2008). Octylphenol, 4-nonylphenol and 4-tert-octylphenol are used in polymeric resins employed in ink preparation (EuPIA, 2012), while nonylphenol is part of some surfactants used in the printing (Miljøstyrelsen, 2011a). The majority of thermal paper is used in cash register receipts, which may contain up to 17,000 mg/kg of BPA (Miljøstyrelsen, 2011b). The remaining chemicals in Table S3 (Supplementary Material) show significantly lower concentration ranges (0.01-68.9 mg/kg) when compared to those of BPA (0.068-17,000 mg/kg).

Liao & Kannan (2011) detected BPA in the majority of 99 paper products analysed, which included magazines, paper towels, napkins, flyers, printing papers, etc., thus indicating potential spreading due to recycling. Similarly, another study (BMELV, 2012) found the highest concentrations of BPA in board packaging which was assumed to have the highest content of recycled paper. Structural BPA analogues (e.g. BPB, BPS, BPF, etc.) are available on the market, but the potential health effects of substitutes are still to be assessed in detail (Rosenmai et al., 2013). Phenols deserve special attention in terms of paper recycling, as nearly all of them demonstrate a high affinity to solids and are persistent, according to biodegradability criteria. The removal of BPA in the deinking process has been observed to be higher than 50%, but this still resulted in average concentrations of BPA of 10 mg/kg in the board produced (BMELV, 2012). This was in contrast to board based on virgin fibres, where no BPA was detected.

3.5 Parabens

Esters of *p*–hydroxybenzoic acid, or parabens, are commonly used as preservatives in a variety of consumer products (Miljøstyrelsen, 2013). Butyl, ethyl, methyl and propyl parabens, identified in Step 2 (Table S4 (Supplementary Material)) and which may be used as preservatives and biocides by both the paper and the printing sectors (Miljøstyrelsen, 2011a; Vinggaard et al., 2000), are all classified as EDCs and show a tendency to remain in aqueous solution. Hence, they can be expected to be removed in the wet end of paper production. Only butyl and propyl parabens show a partial affinity to solids, which may constitute an issue in paper recycling. Although no limit values for chemicals in paper in Table S4 (Supplementary Material) were available, ‘no release of substances in quantities which have an antimicrobial effect’ applies to food-contact paper, in accordance with paper industry guidelines (CEPI, 2012). In a study investigating the oestrogenic potential of paper for household use, parabens (methyl and propyl paraben) were identified only in samples of paper made from virgin fibres (Vinggaard et al., 2000).

3.6 Inorganics

Out of the 157 chemicals, 22 substances were inorganic (Table S5 (Supplementary Material)). Inorganic chemicals in general, and potentially toxic metals in particular, are used mostly in pigment preparation and coatings (Miljøstyrelsen, 2011a). The presence of Hg could not be attributed to any particular process, and it was therefore assumed to be the result of impurities and/or contamination (Huber, 1997). Nevertheless, two studies addressing waste paper composition found Hg in measurable concentrations (Riber et al., 2009; Rotter et al., 2004). Most of the chemicals presented in Table S5 (Supplementary Material) have not been reported based on analytical experiments but rather from inventory lists indicating their use by industry. Concentrations of Hg ranged from 0.01 to 0.386 mg/kg, Cd ranged from 0.02 to 0.3 mg/kg, while total Cr was found in the highest concentrations at between 1.1 and 92 mg/kg of paper. Since some pigments and dyes may contain Pb, one study showed that journals and magazines contained the highest concentrations (up to 400 mg/kg) of Pb in recyclable waste paper (BMELV, 2012). The same study also mentioned that the levels of Hg and Cd found were negligible. The limit values for Cd, Pb and Hg in paper and board intended for use in food packaging were set at 0.5, 3.0 and 0.3 mg/kg, respectively (CEPI, 2012).

Due to the nature of inorganic constituents in waste paper, their removal in the recycling process may vary. One relevant study (BMELV, 2012) indicated that newly produced paper products based on recycled paper may still contain considerable concentrations of Pb (up to 26 mg/kg), while concentrations of some metals (Sn, Sb) may even increase during paper recycling, potentially indicating release from machinery (BMELV, 2012). Nevertheless, the authors of the study indicated that the presence of potentially toxic metals in the concentrations measured should not pose health hazards, even if the paper is to be used for food packaging.

3.7 Other substances

The remaining substances not falling within the previous groups amounted to 67 out of the original 157 (Table S6 (Supplementary Material)). Although data on their identification in paper are scarce, several of the chemicals have been quantified in the scientific literature and reports (BMELV, 2012; Ozaki et al., 2004; Storr-Hansen and Rastogi, 1988; Zheng et al., 2001). Polychlorinated biphenyls (PCBs) are classified as “Persistent Organic Pollutants” and are no longer used in paper production (e.g. in the carbonless copy paper), as they were abolished in 1993 (Breivik et al., 2007). Nevertheless, PCBs may persist in the environment, for example accumulated in trees (Hermanson and Johnson, 2007) or other sources (e.g. books and archives), and they may therefore be introduced into the paper production process. Diisopropyl naphthalene (DIPN) substitutes for PCBs in carbonless copy paper and may be used in other applications (Biedermann and Grob, 2012). It was shown that among the waste paper analysed, office paper contained the highest concentrations of DIPN (up to 1,400 mg/kg), indicating that specialty paper and the use of recycled paper are important sources thereof (BMELV, 2012). The study also showed that unconverted board made from recycled paper and intended for food packaging may contain DIPN ranging from 11 to 27 mg/kg.

Although, since the early 1990s, restrictions in developed countries on the use of elemental chlorine in the paper bleaching process have lowered the possibility of dioxin and furan formation (Ginebreda et al., 2012), these substances may still be detectable in paper products and other papermill outputs, albeit at very low levels (Latorre et al., 2005). The presence of dioxins and furans estimated in papermill effluent waters was in the range of approximately 1-10 ng/m^3 (Latorre et al., 2005), while Munawar et al. identified both in lake sediments near pulp and papermill facilities (Munawar et al., 2000). Another study showed waste paper as the main source of dioxins and furans in a paper recycling facility (Santl et al., 1994). The issue is especially relevant for emerging economies, where potentially more lenient environmental legislations are applied and elemental chlorine may still be in use, thus resulting in detectable levels of dioxins and furans in pulp, paper and effluents (Thacker et al., 2007; Zheng et al., 2001).

The attention of the paper industry to some of the chemicals listed in Table S6 (Supplementary Material) has already been drawn, leading to setting limit concentration values for paper and board used in food packaging: DIPN is subject to tests only in products containing recycled paper, and these concentrations should be ‘as low as technically possible’ (CEPI, 2012). The same guidelines set the limit concentration of Mechler’s ketone as low as 0.0016 mg/dm^2.

3.8 Implications for waste paper recycling and needs for future research

Although a relatively small number of substances were identified as critical (157 out of approximately 10,000), there is a need for more information on their presence in waste paper intended for recycling. Quantitative information on the presence of these substances could provide a basis for establishing a priority list of chemicals to be monitored in waste paper prior to recycling as well as in the final paper products. Although the paper industry has already placed focus on a range of substances (e.g. BPA, BBP), the analytical methods needed to monitor others (e.g. substances constituting mineral oils) are not readily available and represent a challenge for future research. While the specific conditions of the paper recycling processes (i.e. temperature, pH, residence time, etc.) may influence the distribution of chemical substances between the solid, air, and liquid phases, more analyses are needed to fully document substance distributions.

A general lack of transparency related to the use of specific chemicals for example in the printing industry contributes with uncertainty about the substance load associated with paper products and thereby also with the subsequent quality of waste paper as a resource for recycling. Many of the substances screened in this study could not exclusively be associated with paper printing; however, the substances could not be excluded either based on available information.

While banning or gradual phasing out of critical substances in paper production may in the future lead to less chemical substances in paper for recycling, increased source-segregation of individual paper types may also be necessary to ensure a high quality of the paper actually collected for recycling. The preliminary results also indicate the necessity of addressing material quality when establishing target recycling rates. Too high levels of critical substances in waste paper may ultimately mean that this paper should be routed to thermal treatment, thereby enabling the destruction of persistent organic chemicals.

4. Conclusions

The literature review clearly demonstrated that paper and board products, as well as waste paper, may potentially contain a large number of chemical substances, many of these associated with the printing industry. From a total list of 10,000 identified chemicals potentially present in paper products, 157 were classified as hazardous. Fifty-one of these substances were identified as critical as they were likely to remain in the solid matrix during paper recycling and thereby end up in new products based on recycled fibres. The analytical literature reviewed indicated presence of several substances (e.g. phthalates, phenols) in higher concentrations in recycled paper when compared to virgin-fibre based products. If such recycled paper products include food packaging, migration into foodstuff is potentially possible. As almost half of these chemicals (24) are classified as persistent and potentially bio-accumulating, this may pose a risk for consumers. Most of the 51 chemicals are intentionally added during manufacturing, while some of the substances (5) could not be attributed to any of the sectors within the paper industry. These substances may either be added unknowingly by the industry, or originate from contamination of the paper during the use phase or during collection and handling in the waste management phase. The study clearly demonstrates that there is a need for more comprehensive quantitative data documenting the levels of potentially hazard substances in paper sent to recycling as well as the final paper products. Based on the hazard screening procedure, 51 substances have been identified as potentially critical. It is recommended that analytical efforts are directed towards these substances.

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